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oject Fission Products Carried by Lead Sulfate in the Ra La Process.

W. H. Baldwin

J. A. Swartout

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M. D. Peterson





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J. A. Swartout

N. H. Baldwin

March 23, 1948

Fission Products Carried by Lead Sulfate in the Ra La Process

Revived interest in the existing Ra La process (MonN-330) and the desire to increase the output per run by an order of magnitude have recalled to mind a number of omissions in the knowledge of the process. Relatively little was known of the distribution of the fission products in this process. Special interest was expressed in the possibility that volatile fission products might escape that would present problems in handling. Some fission products have been followed through two small scale laboratory runs and have been reported here. More definite information can be obtained from operation in 706-D during the handling of Hanford irradiated slugs.

Experimental Conditions.

Slurry of sulfate precipitate contained

0.93 M UNH plus tracer
0.5 M $HNO_3$
3.4 M $H_2SO_4$
0.3 g. Pb(II)/l as lead nitrate

The solution containing all other substances was heated to 90° C. while 18 M $H_2SO_4$ was added dropwise over 30 minutes. Heating was continued at 90° C. for 1 hour then the slurry was allowed to cool and settle. The cake was washed first with 150 ml 3 M $H_2SO_4$ (per g. Pb(II) used) then 4 times with 150 ml water (per g. Pb(II) used) each time.

The cake was metathesized by heating at 90° C. with 4 M $K_2CO_3$ (24 ml per g. Pb(II) used in sulfate precipitation) for 15 minutes, then diluted with water to 0.5 M $K_2CO_3$, and heating was continued for 30 minutes longer. The metathesis was repeated and the cake was dissolved in 0.5 M $HNO_3$ and diluted to a convenient volume for analysis.

Test run "A" was made on a 50 ml scale in the sulfate precipitation using as tracer 1 ml dissolver solution 1 MA from 706-D, run 30. During the sulfate precipitation, the gases above the vessel were run through 10 ml of 1 to 1 mixture of ethanol and water that was 2 M in NaOH. This solution will be referred to here as the "off-gases".

Test run "B" was made on a 1 liter scale containing 1 ml of dissolver solution in the sulfate precipitation, and the only departure from the general conditions was the use of aerosol (1g/l) in the first water wash of the sulfate cake.

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Discussion of Results.

The analytical data (Courtesy E. I. Wyatt) are contained in tables 1 and 2. In run "A", the percentages are those of the total in the several solutions since a sample was not obtained of the starting solution. Barium yields were 81 and 93% with a large part of the contamination being total rare earths (these include La 140), strontium and cerium. Reynolds and Whitney (NonM-154) reported 83% of the total strontium in the waste from the sulfate precipitation during plant run number 12, while in the runs reported here more Sr accompanied the product.

In run "A", only 0.03% of the original gross beta and gamma was found in the off gases. About 42% of this was iodine most of the remaining beta was not accounted for, but a portion of this discrepancy may be due to difficulties in the analysis for I and Ru.

The product samples (after metathesis) contained small amounts of I and Ru activities. It was felt that Ru might be volatilized from the vessel during the electrolysis step. Since the metathesis products in both runs contained only 0.03% of the starting Ru this might not be hazardous. The product from run "B" is being analyzed further by separation on a Dowex 50 ion-exchange column. Yttrium has been tentatively identified as constituting a small portion of the beta activity. Other constituents have been obtained but not yet identified.

During run "B" it was observed qualitatively that the aerosol added to the wash water used on the sulfate cake made the cake settle in a less dense form and occupy a larger volume. The supernatants decanted after the addition of aerosol were always cloudy.

W. H. Baldwin

W. H. Baldwin

WHB:mh

Distribution:

1. J. A. Swartout
2. W. M. Harty
3. H. W. Stoughton
4. E. J. Witkowski
5. M. D. Peterson
6. W. H. Baldwin
7. Central Files

Table 1

Radiochemical Composition of Solutions

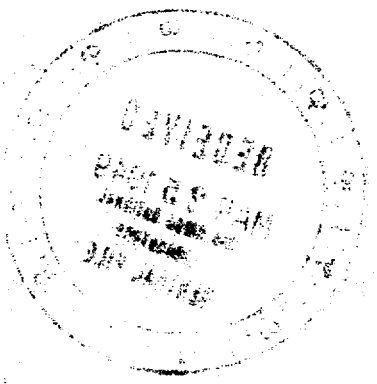
| | Run "A" | | | | Run "B" | |
|---|---|---|---|---|---|---|
| | Sulfate Waste | Off Gases | Metathesis Waste | Product | Starting Solution | Product |
| Solution volume (ml) | 47 | 10 | 25 | 25 | 650 | 50 |
| Gross beta c/m/ml* | 15,500 | 56 | 1,390 | 14,800 | 373 | 1,290 |
| Gross gamma c/m/ml | 27 | 0.1 | 0.65 | 77 | 1.1 | 7.8 |
| Ru beta c/m/ml | 572 | 0.026 | 457 | 0.37 | 15.7 | 0.07 |
| Zr gamma c/m/ml | 11.5 | -- | 0.08 | 0.13 | 0.08 | 0.06 |
| Cb gamma c/m/ml | 9 | -- | <0.002 | <0.002 | 0.1 | 0.007 |
| Ce beta c/m/ml | -- | -- | -- | -- | 44 | 110 |
| Pu alpha c/m/ml | 2.72 | -- | -- | 0.006 | 0.005 | <0.0005 |
| Sr beta c/m/ml | 563 | -- | 230 | 3,600 | 30 | 150 |
| Total Rare Earth beta c/m/ml | 10,300 | -- | 570 | 13,900 | 176 | 600 |
| I beta c/m/ml | 257 | 15. | 49 | 0.04 | 39 | 6 |
| Ba beta c/m/ml | 196 | 0.07 | 26 | 1,640 | 33 | 400 |

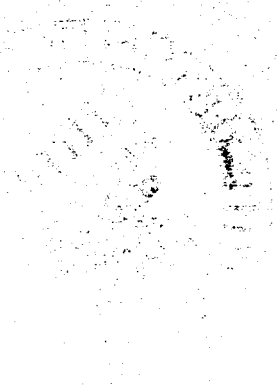
*All c/min/ml have been multiplied by $10^{-4}$.

Table 2

Distribution of Activity

| | Run "A" | | | | Run "B" Product |
|---|---|---|---|---|---|
| | Sulfate Waste | Off Gases | Metathesis Waste | Product | % of that in starting solution |
| Gross beta | 64 | 0.05 | 3 | 33 | 27 |
| Gross gamma | 39.5 | 0.05 | 0.5 | 60 | 55 |
| Ru beta | 61 | 0.01 | 39 | 0.05 | 0.05 |
| Zr gamma | 99 | <0.001 | 0.4 | 0.5 | 5.8 |
| Cb gamma | 100 | — | <0.01 | <0.01 | 0.5 |
| Ce beta | — | — | — | — | 19% |
| Pu alpha | 99.9 | — | — | 0.1 | <0.8% |
| Sr beta | 22 | — | 5 | 73 | 39% |
| Total Rare Earth beta | 57 | — | 2 | 41 | 26 |
| I beta | 90 | 1 | 9 | 0.01 | 1.2 |
| Ba beta | 18 | 0.001 | 1 | 81 | 96 |





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